Thesis SPAM

Nan LIN

Attophysics Group

Monday 16th December 2013, 11h00

L'Orme des Merisiers - Amphithéâtre Claude Bloch (bât. 774)

«APPLICATION OF ATTOSECOND PULSES TO HIGH HARMONIC SPECTROSCOPY OF MOLECULES»

High-order Harmonic Generation (HHG) is an extreme nonlinear process that can be intuitively understood as the sequence of 3 steps: i) tunnel ionization of the target atom/molecule, creating an electronic wave packet (EWP) in the continuum, ii) acceleration of the EWP by the strong laser field and iii) recombination to the core with emission of an attosecond burst of XUV coherent light. HHG thus provides a tunable ultrashort tabletop source of XUV/Soft X-ray radiation on attosecond time scale for applications ('direct' scheme). At the same time, it encodes coherently in the XUV radiation the structure and dynamical charge rearrangement of the radiating atoms/molecules ('self-probing' scheme or High Harmonic Spectroscopy). This thesis is dedicated to both application schemes in attophysics based on advanced characterization and control of the attosecond emission.

In the so-called 'self-probing' scheme, the last step of HHG, the electron-ion re-collision can be considered as a probe process and the emission may encode fruitful information on the recombining system, including molecular structure and dynamics. In the first part, we performed high harmonic spectroscopy of N_2O and CO_2 molecules that are (laser-)aligned with respect to the polarization of the driving laser. We implemented two methods based on optical and quantum interferometry respectively in order to characterize the amplitude and phase of the attosecond emission as a function of both photon energy and alignment angle. We discovered new effects in the high harmonic generation which could not be explained by the structure of the highest occupied molecular orbital (HOMO). Instead, we found that during the interaction with the laser field, two electronic states are coherently excited in the molecular ion and form a hole wave packet moving on an attosecond timescale in the molecule after tunnel ionization. We focused on exploring this coherent electronic motion inside the molecule, and compared the measurements in N_2O and CO_2. The striking difference in the harmonic phase behavior led us to the development of a multi-channel model allowing the extraction of the relative weight and phase of the two channels involved in the emission. An unexpected pi/4 phase shift between the two channels is obtained. Moreover, we studied the attosecond profile of the pulses emitted by these two molecules, and we proposed a simple but flexible way for performing attosecond pulse shaping. In the second part, high harmonic spectroscopy was extended to other molecular systems, including some relatively complex molecules, e.g., SF6 and small hydrocarbons (methane, ethane, ethylene, acetylene). It revealed many interesting results such as phase distortions not previously reported.

For the 'direct' scheme, we photoionized rare gas atoms using well characterized attosecond pulses of XUV coherent radiation combined with an infrared (IR) laser "dressing" field with controlled time delay, stabilized down to about ± 60 as. We evidenced marked differences in the measured angular distributions of the photoelectrons, depending on the number of IR photons exchanged. Joined to a theoretical interpretation, these observations bring new insights into the dynamics of this class of multi-color photoionization processes that are a key step towards studying photoionization in the time domain, with attosecond time resolution.

Vous êtes tous cordialement conviés au pot qui suivra